Thermal metamaterial: Geometric structure, working mechanism, and novel function

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How to freely control heat transfer at macroscopic scale is always a dream of human beings due to important applications in thermal protection, detection, and control/management. Within the past decade (2008-2018), thermal metamaterials have been shown as a promising candidate for this purpose. Here, I review the up-to-date research progress in the field, which mainly includes the following novel phenomena and functional devices together with initial applications: thermal cloak, concentrator, dual function, thermal rotator, macroscopic thermal diode, thermal camouflage, thermal transparency, thermal crystal, energy-free thermostat, abnormal conduction in networks, convection cloak/concentrator/camouflage, and thermal radiation cooling. The underlying mechanisms are presented according to six categories of theory: transformation thermotics, direct solution of Laplace equation, energy band theory, phase transition theory, transformation thermal convection, and thermal radiation theory. I also introduce and comment their prospects from fundamental research to industrial applications. This review has relevance to novel controls of heat transfer (conduction, convection, and radiation) by using artificial structures or devices.

Key words: thermal metamaterial; cloak; concentrator; rotator; diode; camouflage; transparency; thermostat; thermocrystal; radiative cooling

CONTENTS

I. INTRODUCTION

With the advent of energy crisis, high-quality energy sources like coal, oil, natural gas are becoming less and less. However, more and more low-quality energy (say, heat energy) is produced for the inefficient utilization. Therefore, how to make full use of heat energy becomes particularly important. Fortunately, thermal metamaterials come into being as an efficient approach to controlling heat transfer at macroscopic scale.

In fact, heat transfer at microscopic scale has been deeply explored by many scholars, such as Li et al. [1−6], which exerts remarkable influences on this field. However, the macroscopic control of heat transfer is rarely studied.

In 2008, Fan et al. [7] started to develop the transformation optics [8−9] into thermal fields, and predicted the concept of thermal cloak, which has potential applications in thermal protection, misleading infrared detection, and heat preservation or dissipation.

With the establishment of transformation thermotics (or transformation thermodynamics), there comes a research upsurge of achieving novel thermal phenomena via designing artificial structures. The theoretical concept of thermal cloak [7,10−14] has helped to motivate experimental demonstrations [15−19] and popular attention [20,21,22,23].

The so-called "thermal metamaterial" was used by Maldovan in 2013 [24] to name thermal cloaks (shields) and relevant devices designed by using transformation thermotics [7,10,13,15−16], thus causing the formation of the direction of thermal metamaterials. Certainly, the "thermal metamaterial" was only proposed for thermal conduction in 2013 [24], but its connotation has been significantly extended afterwards. As a result, so far, thermal metamaterials also include those artificial structural materials for controlling thermal convection [25−26] and radiation [27−29] with novel properties. In a word, thermal metamaterials can be seen as materials or devices that possess novel macroscopic thermal properties based essentially on their geometrical structures, rather than just physical/thermal properties of component materials [24,30].

In my viewpoint, the existing materials for heat manipulation can be generally classified into two types. One type is dominated by physical properties, such as thermoelectric materials, pyroelectric materials, magnetocaloric materials, photo thermal conversion materials, etc.; the other type is dominated by geometric structures. For the latter, (normal) structural materials can help to realize normal control of heat flow, but thermal metamaterials can be used to realize novel controls (as to be reviewed in this work). So far, the field of thermal metamaterials has aroused enormous research interests, as also evidenced by Google search that shows the search of "thermal metamaterial" occupies 39.3% of all kinds of "metamaterial" as of May 6, 2018.

This review is organized as follows. The main content contains six sections, involving three basic ways of heat transfer: thermal conduction (Sections II–V), thermal convection (Sections VI), and thermal radiation (Sections VII). In Sections II–V, I shall summarize four different methods to control heat conduction.

II. TRANSFORMATION THERMOTICS

In 2008, Fan et al. [7] introduced the theory of transformation optics [8−9] into thermal fields. Together with more new researches [10−13,15], a research direction of transformation thermotics comes to appear. In fact, similar theories of transformation optics have also been introduced into acoustics fields [31], electric fields [32−33], particle diffusion [34], and so on. The subject of this review is to comment on the relevant achievements in thermal fields. Let me start by presenting the first concept predicted by transformation thermotics, namely, thermal cloak.

A. Thermal cloak

As the name of "thermal cloak" suggests, the thermal cloak can be used to hide objects in thermal/temperature fields, as if the objects do not exist.

Fan et al. [7] developed transformation thermotics, and found that although electromagnetic waves and heat fluxes obey different kinds of equations (namely, wave equation versus diffusion equation), they are both form-invariant under coordinate transformation, so the method of transformation optics can be smoothly extended into thermal fields. The transformation thermotics can be mathematically expressed as

$$\kappa' = \frac{J\kappa J^t}{\det(J)}, \tag{1}$$

where κ and κ' are the thermal conductivities before and after transformation, J^t is the transposed matrix of Jacobian transformation matrix J, and $\det(J)$ is the determinant of J. They also found apparent negative thermal conductivities in shaped graded materials [7]. Their work [7] paves a different way to manipulate heat conduction.

Three months later than Fan et al. [7], Chen et al. [10] proposed the modified transformation theory in conduction, and designed a thermal cloak in curvilinearly anisotropic background. Their results show that the theory of transformation thermotics still works, but

the transformed material specifications are position-dependent.

Although the thermal cloak was theoretically predicted as early as 2008 [7,10−12], its experimental validation remains a vacancy. To fill this vacancy, Narayana et al. [15] fabricated the first thermal cloak with multilayer structure (composed of 40 alternating layers of natural latex rubber film and silicone elastomers containing boron nitride particles). Their results bring thermal cloaks from theoretical prediction to practical application, which largely stimulates the follow-up research on thermal cloaks. Incidentally, all the cloaking shells must have thermal conductivities distributed within a gradation profile according to the theory of transformation thermotics, but many experimentalists including Narayana et al. have resorted to multilayer structures for experimentally demonstrating the cloaking behavior. This is physical because the latter are effectively graded according to effective medium theories.
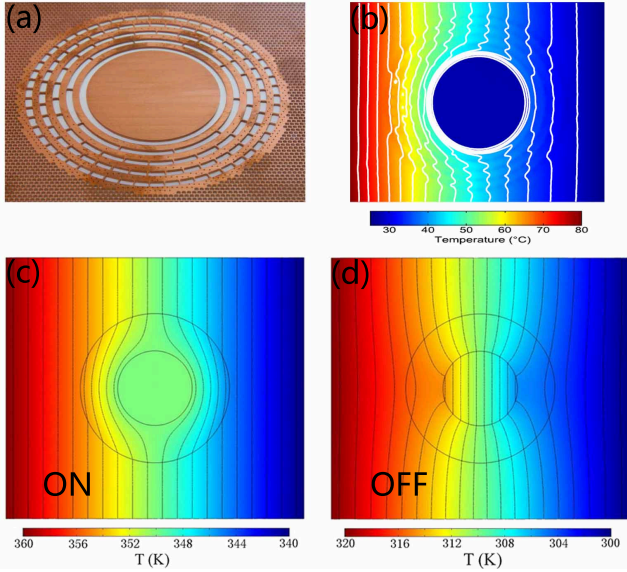

FIG. 1. Thermal cloak. (a, b) Fabricated sample and measured result of transient thermal cloak [16]. (c, d) Simulations of on-state and off-state switchable thermal cloak [36].

However, thermal cloaks theoretically discussed in Refs. [7,10-12] are only suitable for steady states. Guenneau et al. [13] first developed the transformation thermotics to transient (unsteady) states [Fig. 1(a, b)] through considering the density (ρ) and heat capacity (C), namely

$$(\rho C)' = \frac{\rho C}{\det (J)}, \tag{2}$$

which provides an useful method to treat transient domains.

The transient theory also requires experimental validation. Accordingly, Schittny et al. [16] fabricated the first transient thermal cloak, and made transient thermal protection possible.

An underlying limitation is that objects inside thermal cloaks are not sensitive to external field, since the temperature is a constant. To overcome the limitation, Shen et al. [35] theoretically designed a different thermal cloak using transformation thermotics, which made the objects with "feeling" of external fields. However, such scheme requires negative thermal conductivities (which can be realized by adding external work to obey the second law of thermodynamics), which also limits applications.

There is also a limitation that the existing cloaks cannot be switchable (or not intelligent) as ambient temperatures change. Li et al. [36] first proposed the temperature-dependent transformation thermotics, and made the cloak switchable to the change of ambient temperatures. They considered the temperature effect of thermal conductivities. When the temperature is higher than a threshold temperature, the cloak is on [Fig. 1(c)]; when the temperature is lower than the threshold temperature, the cloak is off [Fig. 1(d)]. Their work provides initial guidance on how to design intelligent thermal metamaterials. Here "intelligent" means that the metamaterials can adapt to temperature-changing environments.

Another problem is that the stable temperature of cloaking area will change with the hot/cold sources (which equals to the average temperature of hot/cold sources for symmetric design). To solve this problem, Shen et al. [37] proposed a temperature-trapping method. When the temperature of hot source increases (or the temperature of cold source decreases), phase change will make the thermal conductivity decrease to maintain the original temperature. Such scheme has potential applications in accurate temperature manipulation.

The above-mentioned cloaks are all macroscopic. Ye et al. [38] revealed the mechanism of nanoscale thermal cloaks. They utilized the phonon localization on graphene via chemical functionalization, and gave the molecular dynamics simulations of the cloak. The smallest cloak in the world possesses a different mechanism compared with the macroscopic one, and hence experimental validation remains to be continued.

B. Thermal concentrator

A thermal cloak is to prevent heat fluxes from flowing into it. Another natural thought is to focus heat fluxes (say, thermal concentrator) inside it, which may have potential applications in heat energy collection.

Narayana et al. [15] fabricated the first thermal concentrator by using the effective medium theory. Both
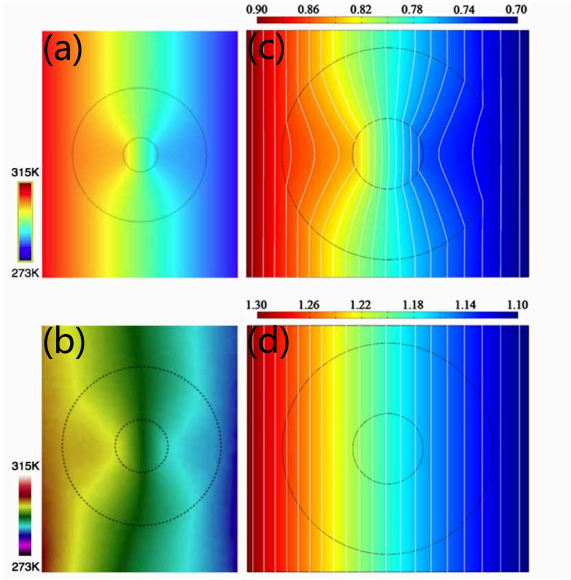
FIG. 2. Thermal concentrator. (a, b) Simulation and experiment of thermal concentrator [15]. (c, d) Simulations of on-state and off-state switchable thermal concentrator [43].

simulation [Fig. 2(a)] and experiment [Fig. 2(b)] are demonstrated to show the validity of their design, which inspires the subsequent research on concentrators.

However, the scheme proposed by Narayana et al. [15] only works in steady states. Therefore, Guenneau et al. [13] extended the scheme into transient (unsteady) states. They gave the theoretical method and related simulations, which laid foundation for treating transient domains.

To further simplify the material parameters, Chen et al. [39] applied the anisotropic coating layer to realize the concentrator. Constant material parameters make the sample fabrication more feasible.

To design practical structures according to the simplified parameters, Wang et al. [40] designed sensu-shaped structure through entropy generation approach. They also defined the concentration ratio, which is useful for better thermal control.

These researches were focused on two dimensions. Han et al. [41] started to design a three-dimensional concentrator with natural-occurring materials, which made the research on thermal concentrators more complete.

To enrich the method of thermal concentrators, Kapadia et al. [42] designed polymeric thermal lens based on the minimization principle of thermal resistance, which is a different method beyond transformation thermotics. Their structure can provide a five-fold enhancement of heat flux, which is of great significance to energy efficiency.

However, all these thermal concentrators cannot be switchable (or not be intelligent). Li et al. [43] utilized the temperature dependence of thermal conductivities to realize the switch of thermal concentrators. Similar to the switchable thermal cloak, when the temperature is lower than a threshold temperature, the concentrator is on [Fig. 2(c)]; when the temperature is higher than the threshold temperature, the concentrator is off [Fig. 2(d)]. This work is instructive, but the parameters are too complicated to realize, at least so far.

Thermal cloaks and thermal concentrators, as two independent phenomena, have been fully studied, which also motivates researchers to study the combination of them to improve practical applications.

C. Bifunctional metamaterials

The first attempt to design bifunctional metamaterials is made by Li et al. [11]. They performed simulations of a bifunctional cloak in both thermal and electric fields. The effective medium theory was applied to design cloak parameters given by transformation approach. This work opens the gate of research on bifunctional metamaterials.

The theoretical prediction by Li et al. [11] requires experimental validation. Ma et al. [17] designed first experiment to realize multiphysics cloaks by directly solving the Laplace equation; see Fig. 3(a, b). Their scheme may extend the research scope on bifunctional metamaterials.

However, the transformation approach can only design single function in single physical domain. Say, Moccia et al. [44] proposed the framework of transformation multiphysics to design thermal concentrator and electric cloak on one device; see Fig. 3(c). Their work may provide guidance for intriguing multifunctional metadevices.

To make the concentrator more applicable, Lan et al. [45] designed the fan-shaped structure to simultaneously realize thermal and electric concentration; see Fig. 3(d, e). Their scheme provides a general method to design bifunctional concentrator.

The above-mentioned bifunctional metamaterials are all designed for different fields (with same/different functions). Shen et al. [46] designed an intelligent cloak-concentrator only on thermal field; see Fig. 3(f, g). They applied the phase change of bimetal sheet to realize the switch between cloak and concentrator. The scheme provides an efficient way to control the temperature gradient.

However, although these research combines the thermal and electric field, they are still two independent transport process. Stedman et al. [47] considered the coupling of thermal and electric field, which means the heat transport will affect the electric-current transport, and vice versa. They developed the transformation
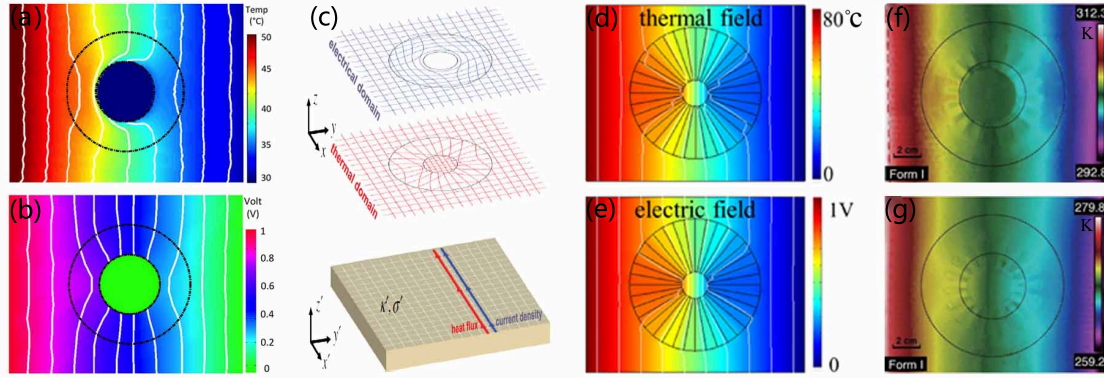
FIG. 3. Multi-functional thermal metamaterials. (a, b) Cloaks for both thermal and electric field [17]. (c) Cloak for electric field and concentrator for thermal field [44]. (d, e) Concentrators for both thermal and electric field [45]. (f, g) Cloak and concentrator for thermal field [46].

principle and realized the cloaking of thermoelectric transport process, which contributes to the manipulation of coupled transport. Beyond the thermal-electric coupling [47], Peng *et al.* [48] studied the thermal-electromagnetic coupling, and designed a chameleonlike cloak which can work under various temperature. The scheme provides a simple way to design unidirectional all-dielectric cloaks.

D. Thermal rotator

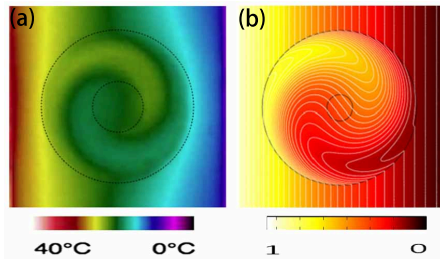Besides thermal cloaks and concentrators, thermal rotators were also proposed to manipulate heat conduction.

FIG. 4. Thermal rotator. (a) is the experimental result for steady states [15], and (b) is the simulation result for transient (unsteady) states [49].

The first thermal rotator [Fig. 4(a)] was fabricated by Narayana *et al.* [15] with the help of effective medium theory. Such scheme has potential applications in misleading thermal detection and reusing heat energy.

However, the concentrator fabricated by Narayana *et al.* [15] only works in steady states. Guenneau *et al.* [49] extended the concentrator into transient (unsteady) domain. They specially designed the density (ρ) and heat capacity (C) of the material, and performed related

simulations [Fig. 4(b)], which makes the research on rotators more complete.

Compared with rotating, refraction is a discontinuous way to change the direction of and thus guide the heat flux. Using thermal metamaterial, heat flux could be bended experimentally by a desired angle and an interesting anomalous refraction of heat flux was observed in [50−52].

E. Applications of thermal cloak, concentrator, and rotator

Sklan *et al.* [53] well concluded the three basic thermal devices (cloak, concentrator, and rotator), and researchers have started to apply these devices to realize practical applications.

Dede *et al.* [54] designed and fabricated a thermal shielding structure on the printed circuit board by utilizing thermal fibre structure of cloak, concentrator, and rotator; see Fig. 5(a). Comparing with the printed circuit board without any modified structure, the optimized fibre structure leads to 10.5 °C temperature reduction; see Fig. 5(b-j). This work opens a gate to apply thermal metamaterials on electronic devices.

Thermal shielding is to prevent heat accumulation. However, heat accumulation may also have positive effect in some situations, especially for the design of thermoelectric generator. Dede *et al.* [55] successfully increased the temperature of hot side by 5.1 °C thanks to optimized thermal fibres and improved the output power by 94.1% in the natural convection system. This technology may be valuable for remote wireless sensing systems with ultra-low power.
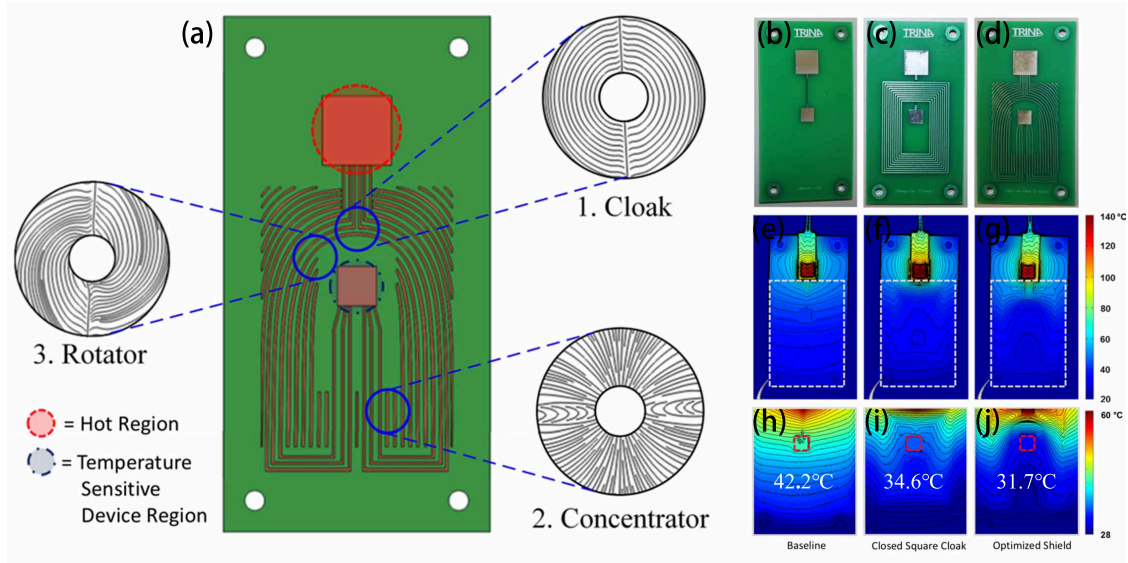
FIG. 5. Thermal shielding [54]. (a) shows how to apply cloak, concentrator and rotator to realize thermal protection. (b–d) Samples of baseline, closed square cloak, and optimized shield. (e–g) are the measured results. (h–j) make the temperature profile of white dashed rectangle in (e–g) more clear. The temperature of red dashed square is 42.2, 34.6, and 31.7 °C, respectively.

F. Thermal diode

Thermal cloaks even have applications in designing thermal diodes. Similar to common electronic diodes, the thermal diodes [1−2] are insulating in one direction [Fig. 6(a)] but conducting [Fig. 6(b)] in the reverse direction. Li et al. [36] first proposed the temperature-dependent transformation thermotics which made a thermal cloak switchable, and then experimentally designed a macroscopic thermal diode; see Fig. 6(c, d). Such scheme has potential applications in heat preservation, heat dissipation, and logical gates. Compared with macroscopic thermal diffusion, microscopic control of thermal diffusion is also of great significance. Loke et al. [56] studied the nanometer-scale, CMOS-integrable, graphene-on-silica stacked materials, and designed thermal-guiding structures for boolean-logic. Their finite-element simulations show that the scheme is useful for advanced (neuromorphic) computation.

In a word, transformation thermotics provides a general and basic method for the effective control of heat conduction. More researches [57−58] are expected to achieve novel heat transfer phenomena.

III. LAPLACE EQUATION

Transformation thermotics provides a revolutionary method to manipulate heat conduction by using the transformation of coordinates. However, the parameters given by transformation thermotics are often
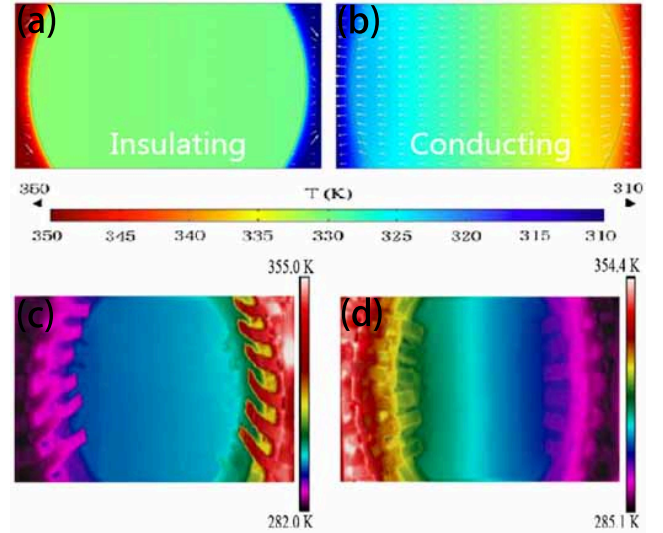

FIG. 6. Thermal diode [36]. (a, b) are the insulating and conducting simulation results, and (c, d) are the corresponding measured results.

anisotropic, nonuniform, and singular, which brings great difficulty to sample fabrication. To solve this problem, researchers pioneered a new way (say, directly solving Laplace equation) to design thermal metamaterials. The key point is to calculate the effective thermal conductivity of the structure by directly solving the governing Laplace equation.
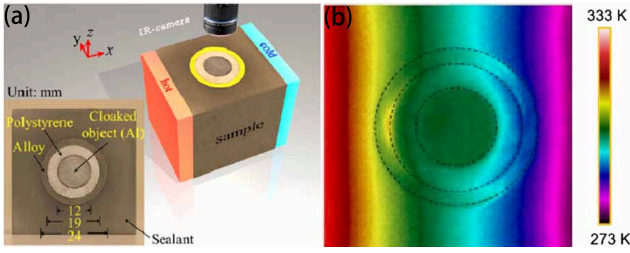
FIG. 7. Bilayer thermal cloak [18]. (a) is the schematic diagram, and (b) is the measured temperature profile.

A. Bilayer thermal cloak

The method based on the direct solution of the Laplace equation was first proposed by Gömöry *et al.* to design magnetic cloaks [59]. Two layers are applied to fabricate the cloak. One is superconductive layer whose permeability (μ) is zero, and the other is ferromagnetic layer (to remove the influence of superconductive layer). Therefore, objects can no longer exert influence on the external field. Such bilayer scheme not only simplifies the material parameters but also acts as an exact cloak.

Similar method was inspirationally introduced into thermotics by Han *et al.* [18], which brings exciting breakthrough to thermal cloak. Bilayer structure is used for sample fabrication [Fig. 7 (a)], which performs well in both steady and transient (unsteady) domains [Fig. 7 (b)]. Compared with thermal cloak designed by transformation thermotics, such bilayer cloak makes thermal invisibility easier to obtain, and contributes to practical applications. Ma *et al.* [17] improved the bilayer structure to fabricate a multiphysical (thermal and electric) field cloak, which is useful for multiphysical manipulation.

The pioneering experiments in Ref. [17-18] were conducted in two dimension. Xu *et al.* [19] extended the scheme into three dimensions, and first fabricated a three-dimensional bilayer thermal cloak. This work opens an avenue to control three-dimensional heat conduction in thermal devices.

B. Thermal camouflage

A thermal cloak is used to hide an object from being detected by external thermal detection. A more expecting function (say, thermal camouflage) is not only to hide an object, but also to disguise the object into another one. This is called thermal camouflage.

Han *et al.* [60] first proposed an innovative scheme of thermal camouflage; see Fig. 8. In their scheme, bilayer cloak is applied to hide the object; see Fig. 8 (a). Then the target object (which arbitrary object is disguised
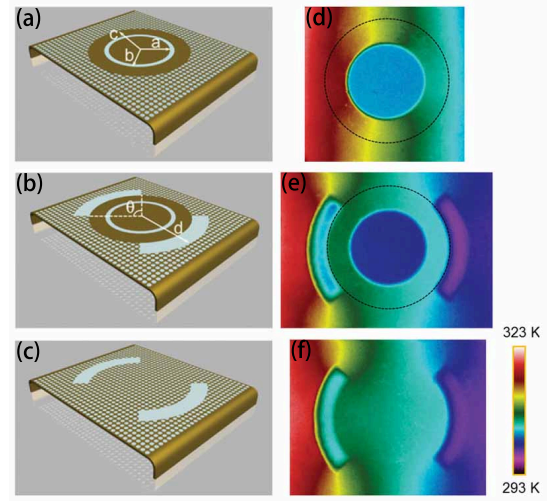

FIG. 8. Thermal camouflage [60]. Bilayer cloak is applied in (a). Target object is applied outside the cloak in (b). (c) is a reference only showing the target object. (d–f) are the corresponding measured temperature profile.

into) is put outside the cloak; see Fig. 8 (b). Therefore, the arbitrary object can be camouflaged into the target one. Thermal camouflage provides potential applications in misleading infrared detection and guidance for designing novel thermal metamaterials.

Such thermal camouflage works perfect in steady states. However, the scheme does not perform well in transient (unsteady) states or with respect to out-of-plane detection. To solve the problem, Yang *et al.* [61] carefully designed the density (ρ) and heat capacity (C) to adapt the transient domain, and applied the neutral inclusion to adapt the out-of-plane detection. Such scheme improves the function of thermal camouflage, and can be more deceptive to thermal detection.

To enrich the phenomena of thermal camouflage, Chen *et al.* [62] designed different camouflage devices through transformation thermotics. Such device can produce the accurate distortion of temperature-distribution profile, but requires negative thermal conductivities (which can be realized by adding external work according to the second law of thermodynamics), which limits potential applications.

The above thermal camouflage [60−62] works for single objects only. Actually the concept of camouflage can also be available for many-particle systems by tailoring many-particle local-field interactions according to effective medium theories [63−64].

The above-mentioned camouflage is only focused on thermal fields. Yang *et al.* [65] extended the camouflage into multiphysical (thermal and electric) fields, where the effective thermal/electric conductivities are carefully calculated. Such scheme may have potential

applications in multiphysical camouflage.

C. Thermal transparency

The Laplace equation also helps to discover electromagnetic wave transparency. Argyropoulos et al. [66] designed the electromagnetic wave transparency, and realize the switch between completely cloaked and strongly resonant. Their scheme is useful for all-optical switches and nanomemories. He et al. [67] introduced the similar concept into thermal conduction. Thermal transparency is achieved by making the effective thermal conductivity of the middle composites equal to that of the background; see Fig. 9 (a). Such scheme has applications in cancelling the so-called thermal stress concentration, which can shorten the service life of materials.

For experimental validation, Zeng et al. [68] designed a heat diffusion device [Fig. 9 (b)] to observe thermal transparency, and validated the theoretical prediction in Ref. [67]. Such device can be applied to thermal protection in chips, satellites, etc. To further simplify the sample structure, Yang et al. [69] designed the single-particle structural materials. The anisotropy of elliptic particle was utilized to realize the manipulation of heat transfer. Their scheme is instructive for designing single-particle structures.
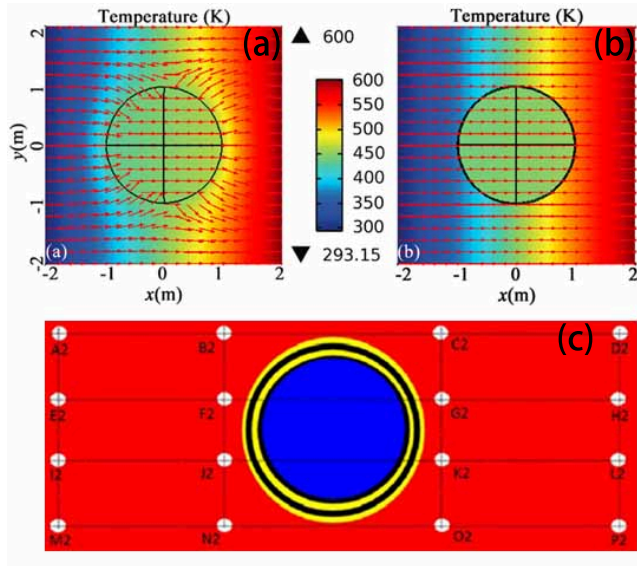

FIG. 9. Thermal transparency. (a, b) Simulation results of thermal transparency [67]. (c) Experimental diagram of thermal transparency [68].

To realize more functions on a single structure, Wang et al. [70] designed the thermal imitators, which is invisible in one direction, and opaque in the perpendicular direction. Such imitators provide guidance for designing thermal metamaterials with different functions in different directions.

To improve practical applications, Xu et al. [71] proposed the periodic structures to realize different functions, including Janus illusion and illusion diodes. Such structures may be helpful in thermal camouflage and rectification.

In a word, compared with transformation thermotics, directly solving Laplace equation provides an efficient way to design thermal metamaterials. Complicated parameters are simplified and novel phenomena are revealed [72,73,74]. More innovative research can be expected in the future with the basic thought of effective medium theories [75,76,77].

IV. MODULATING PHONON BAND STRUCTURE

Photonic [78−79] or phononic [80−81] crystals have been a useful tool in manipulating light or sound waves, respectively. As ionic lattices can affect the motion of electrons, these artificial periodic optical or acoustic structures can also affect photons or phonons, by generating forbidden frequency gaps for corresponding carriers, where Bragg scattering plays an essential role. It is interesting to note that sound and heat both originate from lattice mechanical vibrations and can be described as propagation of phonons from the quantization angle. The main difference is that sound corresponds to low frequencies while the most of heat corresponds to much higher ones (more than THz at room temperature [82]). In other words, phonons carrying sound have a much longer wavelength and behave more like waves at the macroscopic level. Thermal phonons, by contrast, have wavelengths of nanoscale and should be influenced more easily by bulk scattering. If people want to use periodic structures to guide heat like light and sound, the periodicity of such thermocrystal would be too small to be realized in experiments (with a periodicity about 2 nm [82]). What's more, the governing equation for thermal phonons propagation is no longer a wave equation, it is hard to say if there exist explicit band gaps. Luckily, recent advances in modulating phonon band structures have shown potential applications in engineering thermal conductivities and heat management. f

A. Thermocrystals

In response to the difficulties mentioned above, Maldovan [82] developed a method to get a narrow phonon spectrum for heat transfer by shortening the mean-free
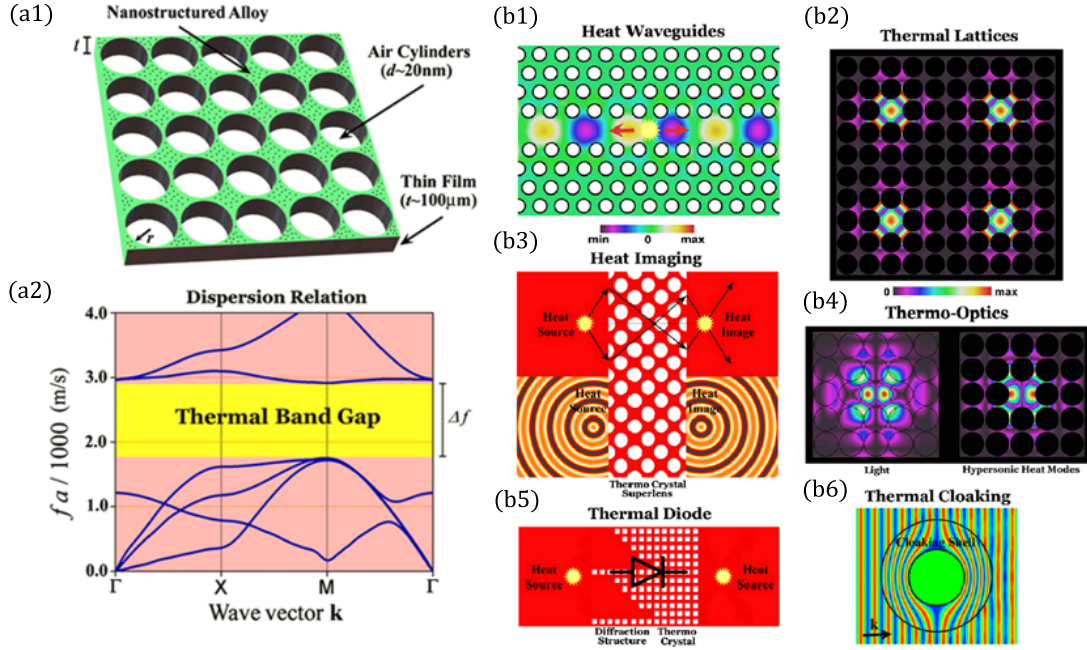
FIG. 10. Thermocrystals [82]. (a1) shows the structure of thermocrystals, where air cylinders are patterned periodically on $Si_{1-x}Ge_x$ alloy film. (a2) shows the phononic band gaps of thermocystals. (b1–b6) are schematic designs for potential applications of thermocrystals, namely, (b1) heat waveguides, (b2) thermal lattices, (b3) heat imaging, (b4) thermo-optics, (b5) thermal diode, and (b6) thermal cloaking.
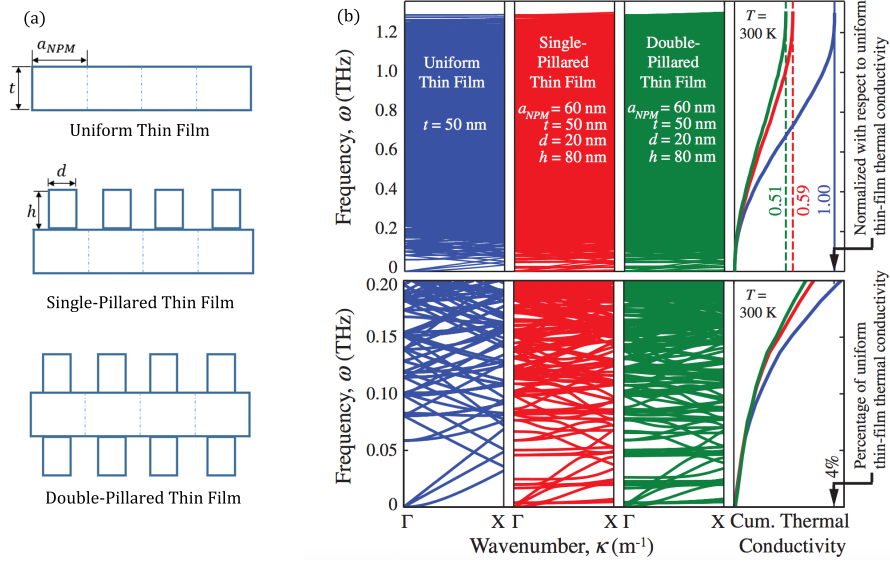

FIG. 11. Nanophononic metamaterial. (a) is the side view of the uniform thin film and single/double-pillared thin film (nanophononic metamaterials). Here t is the thickness of the films, a_{NPM} is the periodicity of square bases in films, d is the side length of a single pillar, which is also a square base and h is the hight of pillars. (b) shows the dispersion relationship for uniform thin film and single/double-pillared thin film, and how the the cumulative thermal conductivity varying with frequency [95].

paths (which is proportional to thermal conductivity) of both high-frequency and very low-frequency phonons. Compared to silicon, Ge nanoparticles are introduced within the $Si_{1-x}Ge_x$ alloy, enhancing mass-difference scattering especially for high-frequency phonons. On the other hand, samples are designed with thin films

so that only very low-frequency phonons could be significantly scattered by rough surface boundaries. Since scattering can reduce phonon mean-free paths, high-frequency and very low-frequency phonons are greatly blocked and a narrowed frequency range of 100–300 GHz contributes most of the thermal conductivity. In this manner, thermal phonons in the $Si_{1-x}Ge_x$ sample can be taken as phonons of elastic waves and methods to fabricate phononic crystals can be applied now. By introducing periodic air holes in the sample (with a periodicity of 10–20 nm), a band gap emerges which matches the specific hypersonic phonon spectrum well (see Fig. 10). This is the first phononic crystal designed to manipulate heat transfer, which can be referred to as thermocrystal. Based on this, various thermal devices with novel functions can be designed (see Fig. 10). With the advance of nanofabrication technologies, thermocrystal could be a revolutionary tool in heat management [24]. It must be noted that, in fact, all the phonons have a greater scattering possibility for adding inclusions and grain boundaries, so thermal conductivities shall be greatly reduced on the whole (also, the reduction of group velocities and density of states play a role, see Ref. [83]), which is important in improving the efficiency of thermoelectric devices [24,84−85].

Many later researches also focus on investigating and exploiting the wave nature of thermal phonons. Anufriev et al. [86] developed a experimental method to manipulate the directions of phonon ballistic transport in silicon membranes where holes arrays were periodically put and proposed a model of thermal lens than can converge phonons at a given point. In other words, they realized thermal guiding and concentrating in nanostructures.

Lee et al. [87] experimentally measured the thermal transport in both periodic (with periodicities not less than 100 nm) and non-periodic nanomeshes and found that phonon backscattering, not phonon coherence, is the main reason why the conductivity is lower than the classical prediction.

Also, after much work had paid attention to how to reduce the thermal conductivity, a layered nanomaterial was proposed to enhance the thermal conductivity of a germanium film by embedding it into two silicon layers to generate interface interaction [88]. What's more, Zhao et al. [89] developed a comprehensive technique tuning the thermal conductivity using selective helium ion irradiation of single Si nanowire. Again to engineer thermal conductivity and manipulate heat transfer, one-dimensional and two-dimensional phononic crystals are investigated to see how phonon interference and bandgaps influence thermal transport in nanostructures with strictly periodicity in [90,91].

B. Hybridization gaps

Different from periodic structure and Bragg-type band gaps, Liu et al. [92] found that the coupling between local resonance and propagating wave is another mechanism for gap formation, which is also called as hybridization gaps [93−94]. Davids and Hussein [95] for the first time proposed a model using hybridization gaps to reduce thermal conductivities, named phononic metamaterial at the nanoscale, where pillars are erected periodically on silicon thin film (see Fig. 11). Every individual pillar is a local resonator, generating standing waves due to total reflection. When the resonant modes and bulk propagating phonon modes converge with the same frequency and polarization, avoided crossing or anti-crossing happens [93−94,96−99], which could flatten the phonon dispersion curves, lower the phonon group velocities, form hybridization band gaps and thus reduce the thermal conductivity (see Fig. 11). Unlike enhancing scattering in nanophononic crystal, hybridization phonon band gaps do not need to introduce defects in materials and wouldn't cause negative influence on the transport of electrons obviously at the same time. Ref. [100-101] later studied how the thermal conductivity of silicon-based nanowires depends on the length of nanowire and the structure of local resonator. Their calculation shows that the phonon free path is also reduced in such phononic metamaterial. This method is especially effective for low frequencies below 4 THz for exploiting the wave nature of phonons while scattering mainly affects phonons at higher frequencies, and these two mechanisms can be combined to get a ultra low thermal conductivity. Since locally resonant phononic crystal can modulate waves whose wavelengths can be much larger than the lattice constant and have good robustness against disorder [92−93,98−99], these researches can provide new hints on thermoelectric conversion. Recently, Zhu et al. [102] found that, different from the coupling of locally resonant and bulk modes, avoided crossings can also happen when random defects introduced in materials.

In short, the advances on modulating phonon band by various nanostructures focus on generate band gaps in low frequencies. Usually the thermal conductivity is reduced, which is important for thermoelectric materials. When the proportion of heat transfer by high-frequency phonons drastically decreases, thermocrystals might be realized. All these methods can greatly improve the ability of controlling heat flow.

V. PHASE TRANSITION THEORY

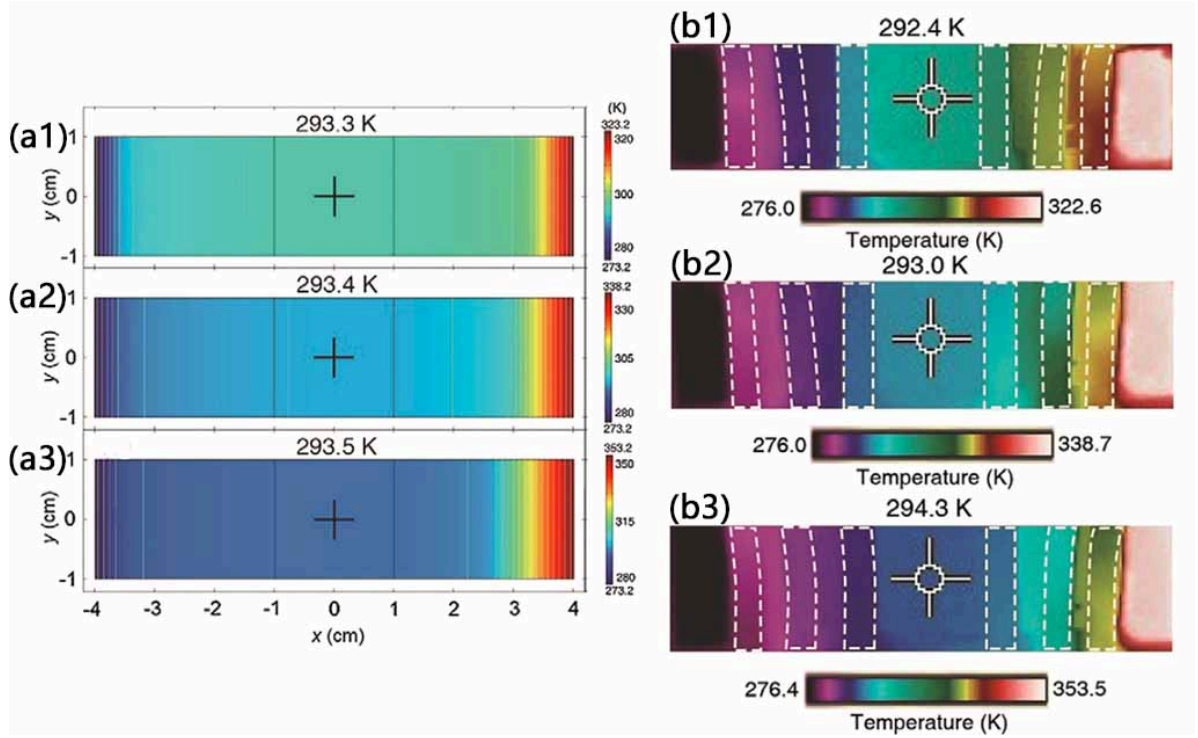Phase transition is a common phenomenon in nature, which is the reason why the world is so wonderful and

FIG. 12. Simulation (a1)–(a3) and experimental (b1)–(b3) verification of thermostat [37]. For simulations, the cold source is kept at 273.2 K, while the hot source is set at (a1) 323.2 K, (a2) 338.2 K, and (a3) 353.2 K, respectively. During this process, the temperature of the central area remains almost unchanged. For experiments, the cold source is kept at 276.0 K, while the hot source is set at (b1) 322.6 K, (b2) 338.7 K, and (b3) 353.5 K, respectively.

colorful. Actually, phase is also useful in manipulating macroscopic heat flux, such as realizing energy-free thermostat, thermal diode, etc. Relevant details can be found in the following.

A. Temperature trapping

Heat preservation is a pretty general need for usual life. Nowadays, it is hard to imagine how to live without air conditioners and refrigerators, which satisfy our needs to keep a specific space at a certain temperature. These tools all require a large amount of energy input. However, as energy crisis has become a more and more serious problem for human beings, it is urgent for scientists to develop new heat preservation mechanism. In order to achieve this goal, Shen *et al.* proposed an energy-free thermostat (associated with environmental temperature gradient) to maintain a certain area as constant temperature even ambient temperature changes [37]. They derive theoretically that one can make a certain area at a constant temperature indeed as long as he/she can find two types of

materials whose thermal conductivities satisfy

$$\kappa_A = L(T_c - T) = \delta + \frac{\varepsilon e^{T-T_c}}{1 + e^{T-T_c}} \qquad (3)$$

$$\kappa_B = L(T - T_c) = \delta + \frac{\varepsilon}{1 + e^{T-T_c}} \qquad (4)$$

where δ is a small value and ε is large enough according to the needs. However, it's hard to find nature materials with thermal conductivities of this form. The researchers design two types of thermal metamaterials with the aid of shape memory alloys. Fig. 12 shows their simulation and experimental results. Keeping the cold source invariant, the center area almost remains the same temperature even if the hot source raises from 323 to 353 K. The experiment results consist well with simulation ones, which indicates that the theory and experiment are satisfactory. This research implies that it is practicable to manufacture thermostat (in environmental temperature gradient) without other energy input. If household appliances, such as air conditioners and refrigerators, do not need energy input any longer, it will save pretty much electric energy. Since most electric energy is still generated from fossil fuels, it will reduce air pollution furthermore.

This two-dimensional system asks the third dimension for help in order to realize the wanted form of

thermal conductivity [37]. This idea was also adopted in the previous study [36]. However, how to realize this form of thermal conductivity for a three-dimensional system still remains to be a challenging project.

B. Thermal network and diode

An electronic diode has been proved to be a great success in controlling electric currents. It lays a foundation for modern information technology. However, the corresponding thermal diode is not paid much attention, and researches about thermal diodes are much fewer. So, it is a natural ideal to apply thermal diodes to manipulate thermal fluxes. In the design of thermal diodes, two key factors are the geometry asymmetry and the nonlinear thermal conductivity. As the first factor is easy to control, the true challenge is how to realize the thermal conductivity with strong nonlinearity. The most common idea is to use the strong nonlinearity of solid-liquid phase transition near critical temperature. However, the thermal conductivity difference is not big enough between solid and liquid phases of a same material. Thus, the rectification ratio are limited. Shang *et al.* recently proposed a kind of thermal metamaterial based on network structure [103], which can experience large jump of thermal conductivity after a switch process. With the aid of this phenomenon, the rectification ratio of thermal diodes can be improved significantly.

As shown in Fig. 13(b1–d1), these three networks are similar. The difference is that (b1) bond width is slightly smaller than node size; (c1) there are no bonds; (d1) bond width equals node size. Thermal conductivity of these three structure are 3.3, 43.0 and 151.1 W/(m·K), respectively. Fig. 13(a2–d6) are experimental and simulation verification of these three values. Comparing Fig. 13(b1) and (d1), the tiny geometry change of bonds causes huge jump of the whole thermal conductivity. Imagine the bonds are composed of material of positive (or negative) thermal expansion coefficient, then one can get a metamaterial whose thermal conductivity changes between 3.3 and 151.1 W/(m·K), i.e., a metamaterial with strong nonlinearity. The critical point is defined as the temperature at which bond width expands (shrinks) to node size.

In the previous studies of thermal conductive networks, researchers mainly focus on discrete element methods, and apply finite element methods to explore macroscopic regular thermal networks [104−106]. Differently, this work [103] not only proposes a potential method to design thermal diodes with a high rectification ratio, but more importantly, brought up an interesting question that why the property of the whole system can be changed so significantly by a tiny

part in the domain of diffusivity equation. This question might interest both scientists in thermotics and networks [107−114]. Despite of the promising effect presented in this work, unfortunately the experimental realization of this thermal diode seems to be difficult.

VI. TRANSFORMING THERMAL CONVECTION

Besides conduction and radiation, convection is another basic form of heat transfer and is usually the dominant one in fluids (namely, liquids or gases). With the movement of liquids or gases, heat can be transferred due to both temperature gradient and bulk flow, which can be described by convection-diffusion equation.

$$\rho_f C_{p,f} \frac{\partial T}{\partial t} + \rho_f C_{p,f}(\vec{v} \cdot \nabla T) = \nabla \cdot (\eta \nabla T) \qquad (5)$$

where T is the temperature, $\vec{v}$ is the velocity distribution of fluid, ρ_f and $C_{p,f}$ are respectively the density and specific heat at constant pressure of fluid material, and η is the thermal conductivity of fluids. In engineering heat-transfer, enhancing convective heat transfer has raised great interest in past decades [115−118]. On the other hand, some works have studied controlling heat when conduction is affected by convection on the boundary by thermal metamaterials [119]. In Ref. [25,120], the transformation theory is firstly applied to handle such convection-diffusion equations of transient states with the form-invariance under coordinate transformation. By transforming the heat conductivity $\eta' = \frac{\mathbf{J}\eta\mathbf{J}^{\mathsf{T}}}{\det \mathbf{J}}$ and velocity $\vec{v}' = \mathbf{J}^{\mathsf{T}}\vec{v}/(\det \mathbf{J})$ according to corresponding geometric transformation, thermal cloaks, concentrators and rotators for convection-diffusion phenomena were designed and also checked by finite element simulation. However, it remained an open problem on how to realize anisotropic thermal conductivities and transformed velocities in fluids.

Inspired by studies on flow control [121], Dai *et al.* [26] proposed a theory of transforming thermal convection for creeping flow in saturated porous media. In their model, equations describing heat and mass transfer of steady states are considered simultaneously:

$$\begin{cases} \rho_f C_{p,f}(\vec{v} \cdot \nabla T) & = \nabla \cdot (\eta_m \nabla T), \\ \nabla p + \frac{\beta}{k}\vec{v} & = 0, \\ \nabla \cdot \vec{v} & = 0, \end{cases} \qquad (6)$$

where k is the permeability, η_m is the effective thermal conductivity of porous media and β denotes the dynamic viscosity. This set of equations, including convection-diffusion equation, the Darcy's law and the equation of continuity, all have form-invariance under coordinate
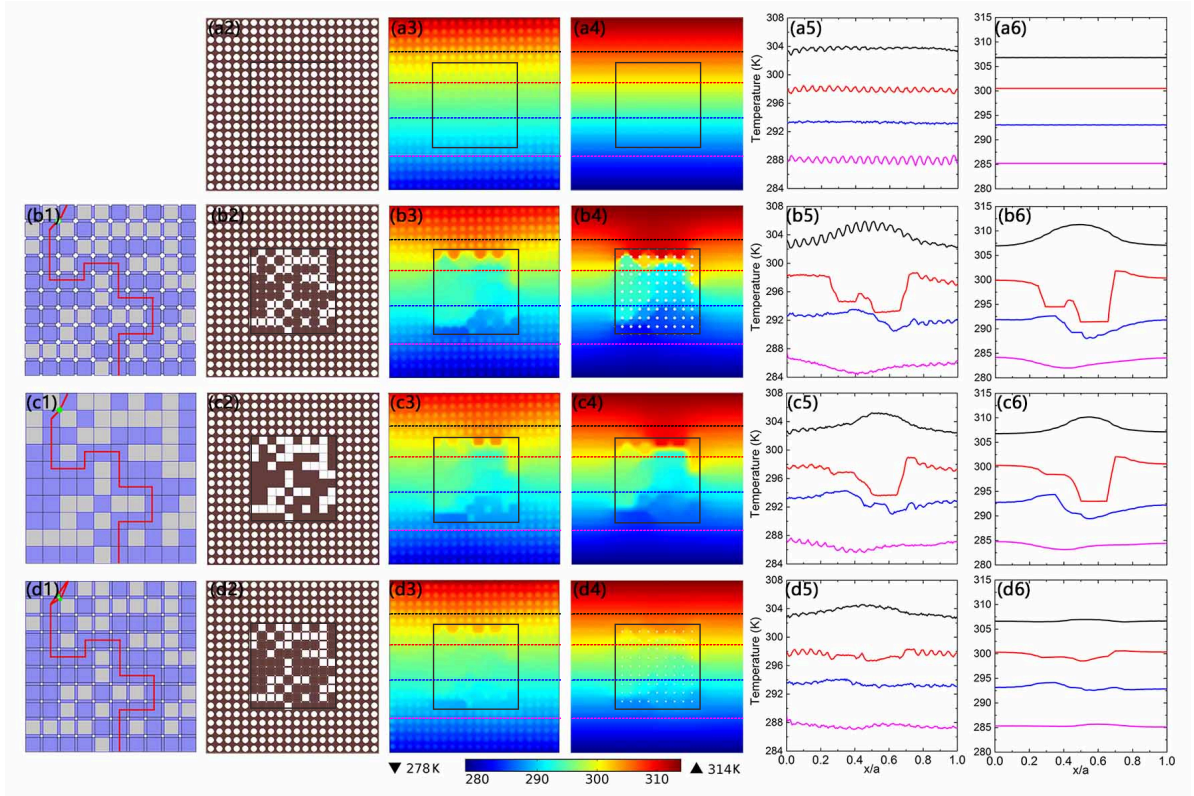
FIG. 13. Thermal conductivity consistency between simulations and experiments [103]. (a2)–(d2) are structure schemes: (a2) reference, (b2) bond width smaller than node size, (c2) no bonds, and (d2) bond width equalling node size. (a3)–(d3) and (a4)–(d4) are corresponding experiment and simulation results. (a5)–(d5) and (a6)–(d6) are temperature distributions extracted from (a3)–(d3) and (a4)–(d4), respectively. (b1)–(d1) shows the explanation for the large thermal conductivity differences caused by tiny geometry changes.

transformation. Again, we should transform the permeability and the thermal conductivity as

$$\begin{cases} \eta'_m = \dfrac{\mathbf{J}\eta_m\mathbf{J}^{\mathsf{T}}}{\det \mathbf{J}}, \\ k' = \dfrac{\mathbf{J}k\mathbf{J}^{\mathsf{T}}}{\det \mathbf{J}}. \end{cases} \tag{7}$$

The transformed permeability facilitates the formation of expected velocity distribution $\vec{v}' = \mathbf{J}^{\mathsf{T}}\vec{v}/(\det \mathbf{J})$ and people can just modulate the conductivity of solid material to get the adjusted effective conductivity. In this way, all the intrinsic properties of fluid material keep the same after transformation, which is beneficial for experimental realization. Based on this theory, controlling heat convection in fluids by metamaterials could be possible.

A. Convection cloaks and concentrators

Refs. [25,120] have theoretically designed thermal cloaks and concentrators with arbitrary shapes based on diffusion-convection equation. In porous media, these concepts are also introduced [26]. The distributions for temperature, velocity and heat flux in the presence or absence of cloaks/concentrators are illustrated in Fig. 14. Simulation results show that such a cloak plays a dual role of thermal cloaking and flow cloaking (see Ref. [121]). The thermal and flow signals are both protected from being scattered by the obstacle placed in the inner core. Also, for a concentrator, it enhances both the speed and heat flux in the inner core. In other words, a bi-functional cloak or concentrator can be realized for thermal and fluid fields.

B. Thermal camouflage in convection

Ref. [26] also considered a simple case of thermal camouflage in convection. The authors put four circles in a background, which is made of a different material. In the existence of a thermal camouflage device, the cloaked object in the inner core behaves just the same as if there were only the four circles when observing the fields of heat or fluid flow. The simulation results are shown in Fig. 15. Such a camouflage device can be used
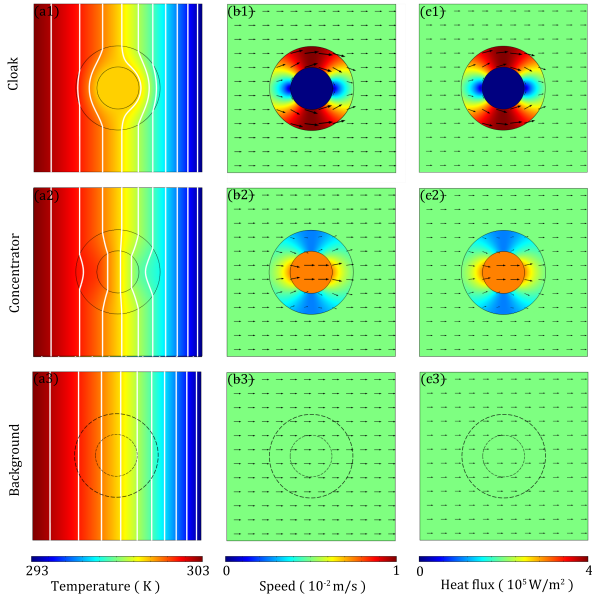
FIG. 14. Simulation results of thermal cloak/concentrator for the x-directed background velocity [26]. (a1), (b1) and (c1) [or (a2), (b2) and (c2)] respectively describe the distributions of temperature, velocity and heat flux in the existence of thermal cloak [or concentrator]. The last line, (a3), (b3) and (c3) describes the temperature, velocity and heat flux where any thermal devices are absent.
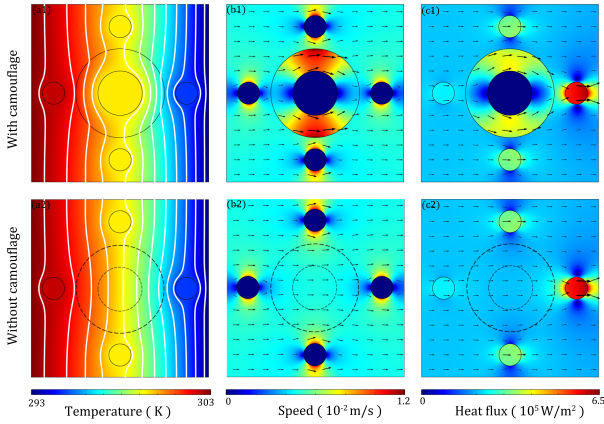

FIG. 15. Simulation results of thermal camouflage when background velocity is along the x-direction [26]. (a1–c1) show the distributions of temperature, velocity and heat flux when camouflage device is put in. Again (a2–c2) show the corresponding distributions as a comparison.

to cheat the infrared detection and the wave detection in fluids.

For the validation of Darcy's law, Ref. [26] only considers a small Reynolds number. When developing further applications, such as concentrators which may help to collect thermal energy from atmospheric circulation,

oceanic circulation, mantle convection, or hydrologic cycle, the Reynolds number can be much higher and some of the governing equations might fail to meet the requirement of transformation theory. Refs. [122-124] studied how to control flow with higher Reynolds number like Brinkman-Stokes flow and small Reynolds number turbulent flow, which might help to advance the research on coupled heat and mass transfer. Also, the whole region in the model of Ref. [26] is filled with porous media and how to get similar effects when the background is free space keeps unclear. In addition, natural convection and transient heat transfer haven't been taken into account yet. This is still a field with many fundamental issues unsolved. Though, the scheme of transforming a coupled series of equations can be useful both for controlling multiple fields interacting with each other (such as in thermoelectric transport [47]) and for managing heat transfer combined with conduction, convection and radiation at the same time.

VII. HEAT RADIATION THEORY

With the development of society and improvement of living standard, air conditioners are used more and more generally to maintain room temperature at a comfortable value in hot summer. In the United States, energy consumption of air conditioners even reaches nearly fifteen percent. Thus, if the energy needed by air conditioners can be reduced, the energy crisis can be eased significantly. Passive cooling system at night has been studied extensively since 1975 and many successful achievements have been gained [125−129]. However, the major energy consumption of air conditioners concentrates at daytime of summer. Thus researchers are struggling to passive cooling methods at daytime [27,130−134]. Among these works, the thermal photonic approach is firstly experimentally realized [27,130,135]. However, this approach needs stringent control in fabrication process, which limits its scalable manufacture significantly. Polymeric photonics approach is considered as a promising method to overcome this shortcoming [131−134,136].

A. Thermal photonic approach

In 2014, Raman *et al.* [27] firstly experimentally realized a passive cooling strategy at daytime via thermal photonic approach. The structure they design is shown in Fig. 16(a). The key part "photonic radiative cooler" is shown as yellow. Fig. 16(b) exhibits its nanostructure.

It's cooling power is given by:

$$P_{\text{cool}}(T) = P_{\text{rad}}(T) - P_{\text{atm}}(T_{\text{amb}}) - P_{\text{Sun}} - P_{\text{cond+conv}}, \quad (8)$$
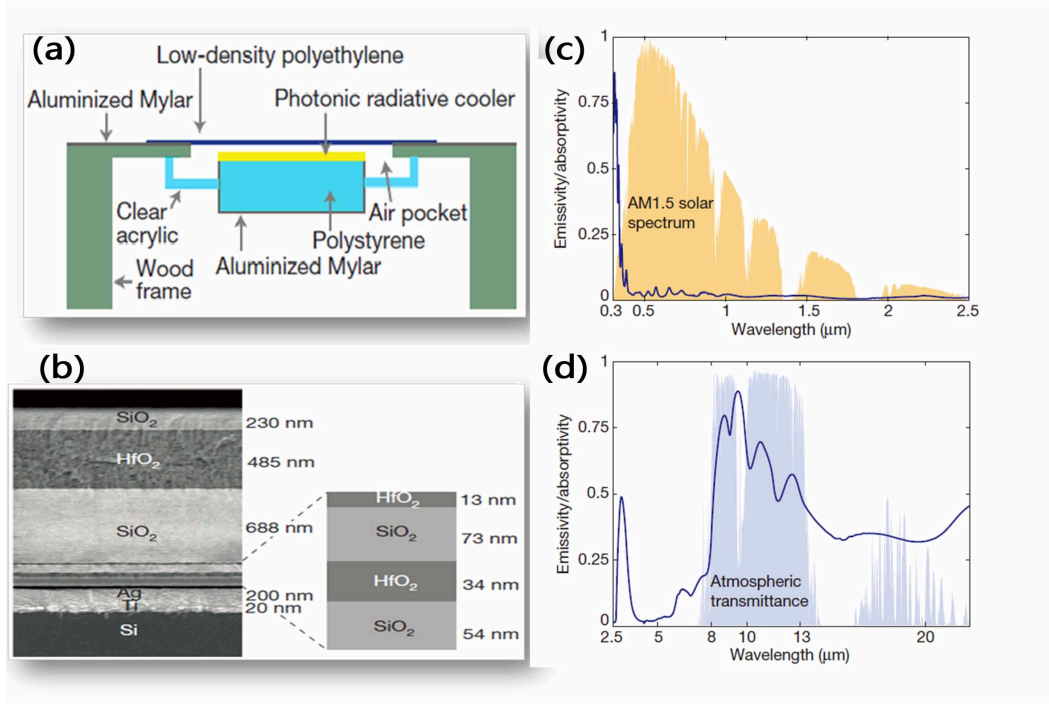
FIG. 16. Structure and principle of Raman's passive photonic radiative cooler [27]. (a) Structure scheme. (b) Nanostructure of the thermal photonic crystal. Blue lines in (c) and (d) denote emissivity/absorptivity of the thermal photonic crystal. Yellow shadow in (c) stands for AM1.5 solar spectrum, and grey shadow in (d) is the atmospheric transmittance.

where, $P_{rad}(T)$ is the power radiated out by the structure, $P_{atm}(T_{amb})$ is the absorbed power due to incident atmospheric thermal radiation, P_{Sun} is the incident solar power absorbed by the structure and $P_{cond+conv}$ is the power lost due to convection and conduction [27]. On one hand, the emissivity/absorptivity spectrum of this structure is designed to be as small as possible at the range of solar spectrum (0.3–2.5 μm), thus P_{Sun} is reduced. On the other hand, the emissivity/absorptivity spectrum is as large as possible at the atmosphere transparent window (8–13 μm), thus the dark universe can be viewed as a cold source and $P_{rad}(T)$ is enhanced largely.

Then their experiment results show that this structure can cool to 4.9 °C below ambient air temperature under the sun. It has a cooling power of 40.1 W/m^2. This work has been a milestone in studying passive cooling at the daytime.

In 2015, Shi et al. [28] conduct a biomimetic research about a kind of silver ant in the Sahara desert. The ant can maintain body temperature in the range of 48–51 ^{0}C even the temperature of desert surface reaches 60–70 °C. The basic principle for the silver to keep 'cool' is reflecting both visible and near-infrared rays and emitting across the atmospheric window. Different from the multilayer structure, the ant emits energy by a dense array of triangular hairs. Besides, the

ants' bare bottom surface reflects most infrared radiation from the desert surface, thus prevent from getting too much heat.
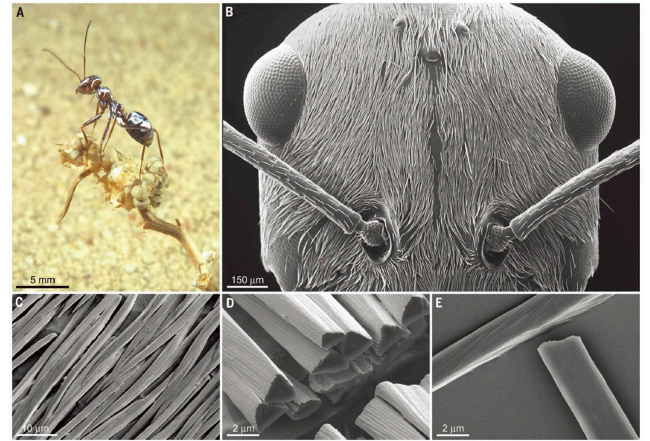

FIG. 17. Silver ant and microstructure of its hairs [28]. (a) A silver ant. (b), (c) and (e)are scanning electron microscopes of the ant's head and hairs. (d) Cross-sectional view of the ant's hair obtained by focused-ion beam.

The principle that the ants 'keep cool' is a good example for researchers to learn from nature and motivate scientists move further in realizing more effective passive cooling.

B. Polymeric photonic approach

Although the multilayer thermal photonic approach has been a great success in passive cooling, it needs strict control of layers' thickness, which implies that it is hard to produce extensively.
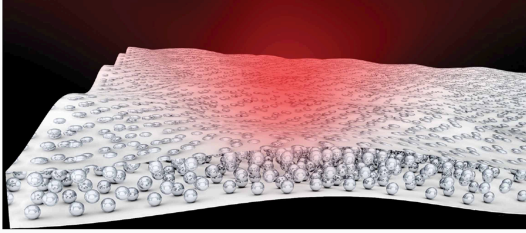

FIG. 18. Glass-polymer hybrid metamaterial for daytime radiative cooling. Spherical SiO_2 particles are embedded in polymethylpentene matrix randomly [29].

In order to make daytime passive cooling technology more efficient and scalable-manufactured, Zhai *et al.* proposed a new metamaterial for daytime radiative cooling which is composed of randomized glass-polymer hybrid [29]. As is shown in Fig. 18, spherical micro SiO_2 particles are distributed in polymethylpentene matrix randomly. After carefully choosing particle size and concentration, the emissivity of this hybrid will exceed 0.93 between 8–13 μm, which is transparency for atmosphere. This hybrid can be manufactured scalably and the researchers produce samples 5 m/min, which is 50 μm in thickness and 300 mm in width. This fact makes it more applicable. However, the thin film is just responsible for emit energy at the range of mid infrared. The function of reflecting visible and near infrared light is in the charge of the silver film covered backward with a thickness of 200 nm.

This work is important not only because it moves a big step to practical application of daytime passive cooling, but also takes two functions apart. The later allows researchers to focus on improving a single target and to avoid much trade-off.

Despite the great success researchers have made, the cooling power is still too small when compared with the solar radiation power (about 900 W/m^2). There is still a long way to go for realizing daytime passive cooling.

C. Radiative camouflage

Moreover, radiation is also applied for camouflage rather than cooling. Li *et al.* [137] first designed a structured thermal surface which can protect arbitrary objects from infrared detection. In their scheme, transformation thermotics and unidirectional thermal cloak are applied to realize their purpose. Their work has practical applications in misleading infrared detections, and may open a new way for exploring thermal radiation.

VIII. SUMMARY AND OUTLOOK

In this review, I have presented both the origin of thermal metamaterials and their main research progress. In general, the concept of thermal metamaterials can help to design various functional materials or devices, which have broad potential applications in thermal protection, detection, and control/management.

Although thermal metamaterials have achieved great attention within the past decade, several key scientific problems remain to be solved. For example, how to control thermal conduction, convection and radiation simultaneously by designing certain thermal metamaterials? How to completely overcome the three limitations associated with transformation thermotics: anisotropic, inhomogeneous, and singular? How to freely tune thermal conductivities via thermal metamaterials, e.g., based on locally resonant nano-structures or periodic nano-structures? The answers to these questions may promote future researches on thermal metamaterials.

Certainly, all the heat-conduction-related researches summarized in this review can be readily extended to other fields like electrostatics, magnetostatics, and particle diffusion, where electric conductivities, magnetic permeabilities and diffusion constants respectively play the same role as thermal conductivities in heat conduction.

Additionally, in the light of nonlinear optics, can one develop its counterpart in thermotics, nonlinear thermotics [138]? Fundamentally, electric permittivities are a function of electric fields in nonlinear optics, and thermal conductivities are a function of temperatures in nonlinear thermotics. Since electric fields (or electric potentials) in nonlinear optics are only mathematically analogous to temperature gradients (or temperatures) in nonlinear thermotics, new physics and applications may be expected in this direction.

Last but not least, to make the research field "thermal metamaterials" survive as long as possible, it is not only necessary to continue the fundamental researches, but also to apply the existing achievements to deal with practical problems. In this direction, a good example is the research on daytime radiative cooling: after Raman *et al.* [27] revealed its mechanism based on a thermal photonic approach, Zhai *et al.* [29] reported high-throughput, economical roll-to-roll manufacturing of such metamaterials, which yields immediate applications. Anyway, more researches for applications are expected in the future.

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热超构材料：几何结构、工作机制与新奇性质

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摘要：因为在热保护、热探测和热管理领域存在重要的应用价值，自由操控宏观热流一直是人类的一个梦想。热超构材料正是为此目的应运而生，它是电磁超构材料在热学领域的延伸。在此，我将综述该领域自 2008 年诞生以来取得的若干研究进展，其将主要包括以下新奇热现象或功能器件：热隐身；热聚集；热旋转；宏观热二极管；热伪装；热透明；热晶体；环境温差中零能耗保温；宏观热网络中反常热传导；热对流隐身、聚集、伪装；热辐射制冷。我将介绍与之相关的微观或宏观传热机制，这些机制可以通过以下理论或方法来理解或阐述：变换热学理论、Laplace 方程、热声子能带理论、相变理论、变换热对流理论、热辐射制冷理论。我也将介绍这些材料从基础研究到工业应用的发展前景。

关键词： 热超构材料；隐身斗篷；聚集器；旋转器；二极管；伪装；透明；恒温器；热晶体；辐射制冷